PATENT COOPERATION TREATY

PCT

INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

(Chapter II of the Patent Cooperation Treaty)

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference PF14L190	**FOR FURTHER ACTION**	See Form PCT/IPEA/416
International application No. PCT/JP2004/018095	International filing date *(day/month/year)* 29.11.2004	Priority date *(day/month/year)* 08.12.2003
International Patent Classification (IPC) or national classification and IPC INV. H01M8/12 H01M4/88 H01M4/94 H01M4/86		
Applicant TOYOTO JIDOSHA KABUSHIKI KAISHA et al.		

1. This report is the international preliminary examination report, established by this International Preliminary Examining Authority under Article 35 and transmitted to the applicant according to Article 36.
2. This REPORT consists of a total of 16 sheets, including this cover sheet.
3. This report is also accompanied by ANNEXES, comprising:
   a. ☒ *sent to the applicant and to the International Bureau)* a total of 6 sheets, as follows:
      ☒ sheets of the description, claims and/or drawings which have been amended and are the basis of this report and/or sheets containing rectifications authorized by this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions).
      ☐ sheets which supersede earlier sheets, but which this Authority considers contain an amendment that goes beyond the disclosure in the international application as filed, as indicated in item 4 of Box No. I and the Supplemental Box.
   b. ☐ *(sent to the International Bureau only)* a total of (indicate type and number of electronic carrier(s)) , containing a sequence listing and/or tables related thereto, in electronic form only, as indicated in the Supplemental Box Relating to Sequence Listing (see Section 802 of the Administrative Instructions).

4. This report contains indications relating to the following items:

- ☒ Box No. I Basis of the report
- ☐ Box No. II Priority
- ☐ Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- ☒ Box No. IV Lack of unity of invention
- ☒ Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- ☐ Box No. VI Certain documents cited
- ☐ Box No. VII Certain defects in the international application
- ☒ Box No. VIII Certain observations on the international application

Date of submission of the demand	Date of completion of this report
27.09.2005	06.03.2006
Name and mailing address of the international preliminary examining authority: European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer Boussard, N Telephone No. +49 89 2399-7196

INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

International application No.
PCT/JP2004/018095

Box No. I Basis of the report

1. With regard to the **language**, this report is based on the international application in the language in which it was filed, unless otherwise indicated under this item.

   ☐ This report is based on translations from the original language into the following language , which is the language of a translation furnished for the purposes of:
   - ☐ international search (under Rules 12.3 and 23.1(b))
   - ☐ publication of the international application (under Rule 12.4)
   - ☐ international preliminary examination (under Rules 55.2 and/or 55.3)

2. With regard to the **elements*** of the international application, this report is based on *(replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report)*:

Description, Pages

1-35	as originally filed

Claims, Numbers

1-20	received on 30.09.2005 with letter of 27.09.2005

Drawings, Sheets

1/10-10/10	as originally filed

☐ a sequence listing and/or any related table(s) - see Supplemental Box Relating to Sequence Listing

3. ☐ The amendments have resulted in the cancellation of:
   - ☐ the description, pages
   - ☐ the claims, Nos.
   - ☐ the drawings, sheets/figs
   - ☐ the sequence listing *(specify)*:
   - ☐ any table(s) related to sequence listing *(specify)*:

4. ☐ This report has been established as if (some of) the amendments annexed to this report and listed below had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).
   - ☐ the description, pages
   - ☐ the claims, Nos.
   - ☐ the drawings, sheets/figs
   - ☐ the sequence listing *(specify)*:
   - ☐ any table(s) related to sequence listing *(specify)*:

* *If item 4 applies, some or all of these sheets may be marked "superseded."*

Box No. IV Lack of unity of invention

1. ☐ In response to the invitation to restrict or pay additional fees, the applicant has:
   - ☐ restricted the claims.
   - ☐ paid additional fees.
   - ☐ paid additional fees under protest.
   - ☐ neither restricted nor paid additional fees.

2. ☒ This Authority found that the requirement of unity of invention is not complied with and chose, according to Rule 68.1, not to invite the applicant to restrict or pay additional fees.

3. This Authority considers that the requirement of unity of invention in accordance with Rules 13.1, 13.2 and 13.3 is

   ☐ complied with.

   ☒ not complied with for the following reasons:

   see separate sheet

4. Consequently, this report has been established in respect of the following parts of the international application:

   ☒ all parts.

   ☐ the parts relating to claims Nos. .

Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes:	Claims	
	No:	Claims	1,2,20
Inventive step (IS)	Yes:	Claims	
	No:	Claims	3-19
Industrial applicability (IA)	Yes:	Claims	1-20
	No:	Claims	

2. Citations and explanations (Rule 70.7):

   see separate sheet

Box No. VIII Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

see separate sheet

Re Item IV

Lack of unity of invention

The separate inventions/groups of inventions are:

1-3, 5 (partly), 20 (partly)

Claims 1-2 disclose a method of manufacturing a fuel cell by forming a thin electrolyte layer having pores on an hydrogen permeable layer, forming a conductive layer having electrical conductivity on the thin electrolyte layer and electronically-discontinued with the hydrogen-permeable layer via the pores.
Claims 3 and 5 disclose a method of forming said conductive layer by releasing a conductive material perpendicularly toward the electrolyte layer.
Claim 20 discloses a fuel cell manufactured according to claims 1-3 and 5.

4, 5 (partly), 20 (partly)

Claims 4-5 disclose a method of forming the conductive layer by releasing a conductive material toward the electrolyte layer at a specific angle.
Claim 20 discloses a fuel cell manufactured according to claims 4-5.

6-9, 20 (partly)

Claims 6-9 disclose a method of forming the conductive layer by first forming a dielectric layer in the pores present in the electrolyte layer, said dielectric layer being made mainly of an insulating material, before coating the electrolyte layer and the dielectric layer with the conductive layer.
Claim 20 discloses a fuel cell manufactured according to claims 6-9.

10-12, 20 (partly)

Claims 10-12 disclose a method of forming the conductive layer by first filling the pores present in the electrolyte layer with fine particles, forming the conductive layer on the electrolyte layer having the pores filled with the fine particles and removing the fine particles from the pores.
Claim 20 discloses a fuel cell manufactured according to claims 10-12.

13-15, 20 (partly)

Claims 13-15 disclose a method of forming the conductive layer by first forming a protective layer to cover the electrolyte layer before forming the conductive layer on the protective layer.
Claim 20 discloses a fuel cell manufactured according to claims 13-15.

16-17, 20 (partly)
Claims 16-17 disclose a method of forming the conductive layer by coating the electrolyte layer with particles of an electrically conductive material having a greater particle diameter than a width of the pores present in the electrolyte layer.
Claim 20 discloses a fuel cell manufactured according to claims 16-17.

18, 20 (partly)
Claim 18 discloses a method of forming the conductive layer by applying a paste, which contains an electrically conductive material and has a predetermined level of viscosity.
Claim 20 discloses a fuel cell manufactured according to claim 18.

19, 20 (partly)
Claim 19 discloses a method of forming the conductive layer by first forming a conductive film of an electrically conductive material before transferring said conductive film onto the electrolyte layer.
Claim 20 discloses a fuel cell manufactured according to claim 19.

They are not so linked as to form a single general inventive concept (Rule 13.1 PCT) for the following reasons:

The common concept linking together the 8 inventions is a manufacturing method of a fuel cell, comprising the steps of forming a thin electrolyte layer having pores on an hydrogen permeable metal layer and then forming a conductive layer on the thin electrolyte layer electronically-discontinued with the hydrogen-permeable metal layer via the pores, wherein the conductive layer has electrical conductivity.

However, **the pores structure in/on the electrolyte is not clearly defined according to the definition of the invention (see description page 1, line 18 and drawings) and the feature "electronically-discontinued [...] via the pores" is not clear. Therefore the**

common concept cannot be distinguished over the prior art and is still already known from document US2002/0028367, which discloses (page 7, §81 and page 8, §87) a method of manufacturing a fuel cell comprising the steps of forming a porous electrolyte layer on a Pd anode and then spraying a layer of Pt as cathode on said formed electrolyte layer.
Moreover any fuel cell manufacturing method intends implicitly to prevent a potential short-circuit by avoiding an electrical connection between anode and electrode, and the hydrogen permeable metal layer and the conductive layer can act as anode and cathode respectively (see description on page 12, lines 4-5 and page 14, lines 12-13).

The essential feature of the first invention is the formation of the conductive layer by releasing a conductive material toward the electrolyte layer in a direction perpendicular to the electrolyte layer to form the conductive layer thinner than the electrolyte layer. The problem solved is to make the conductive layer formed on the electrolyte layer discrete from the conductive layer of the electrically conductive material formed inside the pores of the electrolyte layer (see description on page 3, lines 10-13).

The essential feature of the second invention is the formation of the conductive layer by releasing a conductive material toward the electrolyte layer at a specific angle. The problem solved is the prevention of the deposition of the electrically conductive material on the hydrogen permeable metal layer exposed on the pores in the electrolyte layer (see description from page 3, line 25 to page 4, line 1).

The essential feature of the third invention is the formation of the conductive layer by first forming a dielectric layer in the pores present in the electrolyte layer, said dielectric layer being made mainly of an insulating material, before coating the electrolyte layer and the dielectric layer with the conductive layer. The problem solved is to block off a connection between surface of the hydrogen metal layer, which is exposed on the pores present in the electrolyte layer, and outside of the pores (see description on page 5, lines 5-7).

The essential feature of the fourth invention is the formation of the conductive layer by first filling the pores present in the electrolyte layer with fine particles, forming the conductive layer on the electrolyte layer having the pores filled with the fine particles and removing the fine particles from the pores. The problem solved is to remove the part of the conductive

layer covering over the fine particles in the pores while removing the fine particles from the pores of the electrolyte layer, thereby enhancing the reliability of insulation between the conductive layer and the hydrogen permeable metal layer (see description on page 6, lines 10-14).

The essential feature of the fifth invention is the formation of the conductive layer by first forming a protective layer to cover the electrolyte layer before forming the conductive layer on the protective layer. The problem solved is to prevent the electrically conductive material from entering the pores of the electrolyte layer in the course of formation of the conductive layer (see description on page 7, lines 3-5).

The essential feature of the sixth invention is the formation of the conductive layer by coating the electrolyte layer with particles of an electrically conductive material having a greater particle diameter than a width of the pores present in the electrolyte layer. The problem solved is to prevent the electrically conductive material from entering the pores of the electrolyte layer (see description on page 8, lines 2-3).

The essential feature of the seventh invention is the formation of the conductive layer by applying a paste, which contains an electrically conductive material and has a predetermined level of viscosity. The problem solved is to prevent the paste containing the electrically conductive material from entering the pores of the electrolyte layer by regulating the viscosity of the paste (see description on page 8, lines 17-21).

The essential feature of the eighth invention is the formation of the conductive layer by first forming a conductive film of an electrically conductive material before transferring said conductive film onto the electrolyte layer. The problem solved is the improvement of the mutual bonding power of the particles of the electrically conductive material, thus preventing the electrically conductive material from entering the pores of the electrolyte layer in the process of transferring the conductive film onto the electrolyte layer (see description on page 9, lines 4-8).

Re Item V

Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Reference is made to the following documents:

D1: US 2002/028367 A1 (SAMMES NIGEL ET AL) 7 March 2002 (2002-03-07)
D2: US-A-5 753 385 (JANKOWSKI ET AL) 19 May 1998 (1998-05-19)
D3: US-B1-6 645 656 (CHEN XIN ET AL) 11 November 2003 (2003-11-11)
D4: US-A-5 741 406 (BARNETT ET AL) 21 April 1998 (1998-04-21)
D5: US-A-4 963 239 (SHIMAMURA ET AL) 16 October 1990 (1990-10-16)
D6: US-B1-6 197 167 (TANAKA YOICHIRO) 6 March 2001 (2001-03-06)
D7: US-A-5 656 387 (BARNETT ET AL) 12 August 1997 (1997-08-12)
D8: US-B1-6 428 920 (BADDING MICHAEL E ET AL) 6 August 2002 (2002-08-06)
D9: US-A-5 543 239 (VIRKAR ET AL) 6 August 1996 (1996-08-06)
D10: US-B1-6 387 230 (MURPHY OLIVER J ET AL) 14 May 2002 (2002-05-14)
D11: PATENT ABSTRACTS OF JAPAN vol. 016, no. 469 (E-1271), 29 September 1992 (1992-09-29) & JP 04

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D12: EP-A-1 258 283 (TOYOTA JIDOSHA KABUSHIKI KAISHA) 20 November 2002 (2002-11-20)

D13: US 2003/059658 A1 (KOHLER JOACHIM ET AL) 27 March 2003 (2003-03-27)

D14: US-A-6 066 364 (BLASS ET AL) 23 May 2000 (2000-05-23)

D15: EP-A-0 621 648 (C. UYEMURA & CO, LTD; UEMURA KOGYO KK) 26 October 1994 (1994-10-26)

D16: US-A-5 415 888 (BANERJEE ET AL) 16 May 1995 (1995-05-16)

D17: US 2002/055034 A1 (FUKUDA KAORU ET AL) 9 May 2002 (2002-05-09)

D18: US-B1-6 475 249 (HSU CHENG HSIEN ET AL) 5 November 2002 (2002-11-05)

D19: US 2003/091890 A1 (FUKUDA KAORU ET AL) 15 May 2003 (2003-05-15)

2. Novelty - Article 33(2) PCT

The present application does not meet the criteria of Article 33(1) PCT, because the subject-matter of claims 1, 2 and 20 is not novel.

2.1 Document D1 discloses (page 7, §81 and page 8, §87) a method of manufacturing a fuel cell comprising the steps of forming a porous electrolyte layer on a Pd anode and then spraying a layer of Pt as cathode on said formed electrolyte layer.

According to the clarity objections raised in Item VIII, the subject-matter of claim 1 cannot be distinguished over the prior art and the manufacturing method known from D1 meets consequently the wording of claim 1.
Moreover any fuel cell manufacturing method intends implicitly to prevent a potential short-circuit by avoiding an electrical connection between anode and electrode. In addition said hydrogen permeable metal layer and said conductive layer can act as anode and cathode respectively (see description on page 12, lines 4-5 and page 14, lines 12-13).

Consequently claims 1 and 2 lack novelty with regard to document D1.

2.2 Claim 20 is a product claim 'per se' and discloses a fuel cell, which is defined by the steps of its manufacture rather than by its own technical features. Said fuel cell has no particularly or unexpected properties involved by its manufacture process since any fuel cell manufacturing method intends implicitly to prevent a potential short-circuit by avoiding an electrical connection between anode and electrode. Said product cannot therefore be distinguished over the prior art. Therefore any document which discloses such a claimed product - even if obtained by a different process or if used for a different purpose - takes away the novelty of the claimed subject-matter. Consequently claim 20 lacks novelty with regard to e.g. document D1.

3. Inventive step - Article 33(3) PCT

The subject-matter of claims 3-19 differs from D1 in the way of forming the conductive layer onto the porous electrolyte. The objective problems solved by the 8 methods of forming the conductive layer claimed in claims 3-19 are mentioned in item IV.

3.1 The present application does not meet the criteria of Article 33(1) PCT, because the subject-matter of claims 3 and 5 does not involve an inventive step.

It is namely well known from the prior art (see e.g. D2 (col 5, lines 19-25; col 7, lines 13-14) or D3 (col 5, lines 17-40; fig 1)) to deposit a cathode, i.e. a conductive layer, onto an electrolyte layer by vacuum techniques using a projection direction substantially perpendicular to the substrate and forming an electrode layer thinner than the electrolyte layer.

3.2 Any fuel cell manufacturing method intends implicitly to prevent a potential short-circuit by avoiding an electrical connection between anode and electrode. Moreover conventional sputtering processes lead generally to a poor or to a lack of pore bottom coverage, which further depends on the pore dimensions, ie aspect ratio (see D4 (col 3, line 59 to col 4, line 19), D5 (col 2, line 53 to col 3, line 4), D6 (col 1, lines 55-67; col 2, lines 7-13, lines 46-49) or D7 (col 3, lines 58-61). The claimed conditions of claim 4 are thus implicitly fulfilled when applying conventional sputtering processes (e.g. with pores having high aspect ratios or without added bias) for depositing a cathode onto the porous electrolyte layer known from D1. The specific angle in documents D4-D6 is 90° and this angle is variable in D7.

Claims 4 and 5 therefore are not inventive.

3.3 The present application does not meet the criteria of Article 33(1) PCT, because the subject-matter of claims 6-9 does not involve an inventive step.

Documents D8 (col 2, lines 59-64; col 4, lines 49-60) and D9 (col 2, lines 20-40; col 3, lines 57-59) both disclose the deposition of a dielectric and insulating layer on the surface of the electrolyte before the deposition of the electrode/catalyst layer thereon. Eventual pores at the surface of the electrolyte therefore are implicitly filled by said dielectric layer and short-circuit is prevented.
Claims 6 and 8 therefore are not inventive.

Document D10 discloses (col 8, lines 46-51; col 9, lines 12-16) a porous polymer membrane filled with inorganic oxide particles in the pores. Claims 6 and 7 therefore are not inventive.

The additional feature of dependent claim 9 does not make claims 1 and 6 inventive,

since the metal oxidation of a metal to form a dielectric material is well-known from the prior art (see document D11).

3.4 Claims 10-12 are considered to be novel and inventive, since none of the cited documents D8 to D10 and D12 discloses or suggests a method comprising the steps of filling the pores of the electrolyte with fine particles and removing said fine particles subsequent to the forming of a conductive layer thereon.

Indeed, documents D8 (col 2, lines 59-64; col 4, lines 49-60) and D9 (col 2, lines 20-40; col 3, lines 57-59) disclose the deposition of a dielectric and insulating particulate material on the surface of the electrolyte before the deposition of the electrode/catalyst layer thereon, document D10 discloses (col 8, lines 46-51; col 9, lines 12-16) a porous polymer membrane filled with inorganic oxide particles in the pores and document D12 (col 7, lines 8-20) discloses a process for forming a porous ceramic support medium and filling the pores of said support medium with metal particles . No removing step of the particles is disclosed in documents 1-4.

3.5 The present application does not meet the criteria of Article 33(1) PCT, because the subject-matter of claims 13 and 15 does not involve an inventive step.

Documents D8 (col 2, lines 59-64; col 4, lines 49-60) and D9 (col 2, lines 20-40; col 3, lines 57-59) both disclose the deposition of a dielectric and insulating layer on the surface of the electrolyte before the deposition of the electrode/catalyst layer thereon. Said dielectric layer may be of the same material as the electrolyte layer. The electrode/catalyst material is therefore prevented from entering the pores of the electrolyte layer.

Claims 13 and 15 therefore are not inventive.

Document D13 discloses (page 1, §8) a method of coating an electrolyte layer with a protective film before coating said protective film with a catalyst ink containing Pt. The protective film is then pulled off. The catalyst material is therefore prevented from entering the pores of the electrolyte layer.

Claims 13 and 14 therefore are not inventive.

3.6 **Claims 16 and 17 are novel and inventive**, since none of the cited documents discloses or suggests a method of forming the conductive layer on the electrolyte layer using electrically conductive particles having a greater particle diameter than a width of the pores present in the electrolyte layer.

Indeed, document D14 discloses (col 1, lines 36-42; col 2, lines 57-61) a method for forming an electrolyte layer onto a porous substrate by pouring a suspension of coarse and also fine electrolyte particles onto the substrate for clogging the pores of the substrate. The so formed structure is then dried and sintered.

Document D15 discloses (page 3, lines 50-54) an arc ion plating method for depositing a metal layer onto a network plastic surface on its entire lattice surface.

3.7 The present application does not meet the criteria of Article 33(1) PCT, because the subject-matter of claim 18 does not involve an inventive step. Indeed document D16 discloses (col 5, lines 63-67; col 6, lines 7-14) a method of coating an electrode ink onto an electrolyte membrane without interacting with the polymers contained in the membrane by adjusting the viscosity to e.g. 10^2poises, i.e. at a relatively high viscosity value. The claimed conditions disclosed in claim 18 are thus implicitly fulfilled and pores invasion is prevented. Claim 18 is consequently not inventive.

Claim 18 lacks also inventive step with regard to document D17 (page 2, §27 and 33) for the same reasons as above-mentioned (viscosity of e.g. 250poises).

3.8 The present application does not meet the criteria of Article 33(1) PCT, because the subject-matter of claim 19 does not involve an inventive step. Indeed document D18 discloses (col 2, lines 54-57; col 4, lines 11-12) a method of coating a catalyst slurry onto a transfer paper and then transferring the catalyst layer (e.g. Pt/C) to an electrolyte membrane by hot-pressing. The catalyst material is therefore prevented from entering the pores of the electrolyte layer.

Moreover document D19 discloses (page 2, §24 to page 3, §25) a method of

fabricating an electrode sheet comprising platinum, transferring it onto the surface of an electrolyte membrane and joining the 2 layers by thermo-compression moulding. The catalyst material is therefore prevented from entering the pores of the electrolyte layer. Claim 19 therefore is not inventive.

Re Item VIII

Certain observations on the international application

1. The feature "electronically-discontinued [...] via the pores" of claim 1 is not clear, thereby rendering the definition of the subject-matter of claim 1 unclear, Article 6 PCT.
   Moreover the pores structure in/on the electrolyte is not clearly defined according to the definition of the invention (see description page 1, line 18 and drawings).

2. The term "thin" used througout the claims is vague and unclear and leaves the reader in doubt as to the meaning of the technical features to which it refers, thereby rendering the definition of the subject-matter of the claims unclear, Article 6 PCT.